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Superheavy Elements

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Chapter 10

Superheavy Elements

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Summary: The long quest to detect Superheavy Elements (SHEs) that might exist in nature and the efforts to artificially synthesize them at accelerators or in multiple-neutron capture reactions is briefly reviewed. Recent reports of the production and detection of the SHEs 113, 114, 115, 116, and 118 are summarized and discussed. Implications of these discoveries and the prospects for the existence and discovery of additional SHE species are considered.

1. EARLY PREDICTIONS OF SUPERHEAVY ELEMENTS

The possibility of relatively stable elements well beyond uranium, the heaviest element found in large quantities in nature, was considered in the early 1950s. This interest was sparked by the totally unexpected discovery of the new elements 99 (einsteinium) and 100 (fermium) in debris from the first U.S. thermonuclear device 'Mike', tested on Eniwetok Atoll in the South Pacific on November 1, 1952 by the Los Alamos Scientific Laboratory. Prior to that discovery, only the elements through californium (atomic number 98) were known. Scientists postulated that the enormous, nearly instantaneous high neutron flux generated in the 10-megaton detonation of Mike resulted in the successive capture of at least 17 neutrons in the uranium-238 (^{238}U) present in the device. In this way, the heavier uranium isotopes through ^{255}U were produced and many of these isotopes decayed rapidly by successive emission of negatively charged β particles to produce isotopes of known elements with atomic numbers of 93 through 98. The uranium isotopes of masses 253 and

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255 decayed all the way to the new elements having proton numbers of 99 and 100 and mass numbers of 253 and 255 as shown in the schematic diagram in FIGURE 1.

Publication (Ghiorso *et al.* 1955) of these 1952-53 discoveries of elements 99 and 100 by Berkeley, Argonne and Los Alamos scientists was delayed until after the data were declassified in 1955. During that same year, John A. Wheeler published a paper (Wheeler 1955) on nuclear fission and nuclear stability in which he extrapolated the rates of spontaneous fission (SF) and other processes limiting nuclear stability to the region of very large masses. In a talk presented at the 1955 International Conference on the Peaceful Uses of Atomic Energy (Wheeler 1956) he showed a diagram of the estimated limits of nuclear stability within which half-lives would be greater than a ten-thousandth of a second. Although he cautioned that his extrapolations might be appreciably in error, he still concluded that it was reasonable to look for nuclei with masses perhaps twice as heavy as ^{256}100, the heaviest nucleus known at the time, i.e., masses of 500 or more! He further suggested that massive neutron irradiation of existing heavy nuclides might be an appropriate method for building such superheavy nuclei.

SUPERHEAVY ELEMENTS - FIGURE 1

FIGURE 1. Schematic diagram of production of heavy uranium isotopes by successive neutron captures in ^{238}U followed by their subsequent β decay to spontaneously fissioning or α-decaying nuclides. Mass chains detected in debris from the Mike thermonuclear test are shown.

The possible existence of superheavy elements was also discussed by Gertrude Scharff-Goldhaber (Scharff-Goldhaber 1957). She speculated that there might be another region of relative stability around ^{310}126 because it might be expected to have two especially stable, spherical closed nuclear shells (similar to closed electron shells). These spherical shells, or 'magic' numbers as they are often called, were thought to be at proton number 126 and neutron number 184, thus making ^{310}126 a 'doubly magic' nucleus.

Such ideas spurred the quest to produce still heavier elements in subsequent thermonuclear tests. Indeed, the rather long-lived nuclide ^{257}Fm (half-life ($T_{1/2}$) = 100 days) was detected in later nuclear tests, indicating capture of at least 19 neutrons in uranium. However, attempts to produce and detect still heavier elements in underground nuclear tests conducted at the Nevada Test Site all failed, thus dashing hopes that heavier long-lived elements could be produced via this multiple neutron-capture process that, as it had been postulated, might 'mimic' production of heavy elements in astrophysical processes. (See Chapter 1 in this Volume.) These unsuccessful attempts were reviewed by R. W. Hoff (1978) at a symposium held at Berkeley in 1978 to commemorate the 25th anniversary of the discovery of elements 99 and 100. He postulated that the neutron fluxes were high enough to have produced masses heavier than 257, but the measured SF half-lives of 0.4 ms for ^{258}Fm and 1.5 s for ^{259}Fm were so short that formation of masses heavier than 257 by

additional neutron capture was effectively blocked. It was suggested that perhaps the very neutron-rich, long-lived ($T_{1/2} \approx 9700$ years) nuclide, ^{250}Cm, formed in rather large quantities in these tests, might be recovered afterward. It could then be used as target material to produce superheavy elements (hereafter to be called SHEs) by bombardments at accelerators with projectiles as heavy as ^{238}U.

Myers and Swiatecki (1966) and Meldner (1967) predicted that an 'Island of Superheavy Elements' well beyond uranium might exist around elements with atomic numbers 114 or 126. This raised the possibility that very long-lived SHEs might still exist on earth after having been formed during the last nucleosynthesis in our solar system some 5 billion years ago. Later theoretical studies based on new theories of nuclear structure (Strutinsky 1966, Nilsson *et al.* 1969a, Fiset and Nix 1972, Randrup *et al.* 1974) confirmed that an island of nuclear stability stabilized by spherical nuclear shells should be centered around 110 to 114 protons and 184 neutrons. Such calculations led to the conclusion that these spherical closed nuclear shells or 'magic numbers' should be nearly as strong as those at 82 protons and 126 neutrons found in doubly magic, stable (non-radioactive) lead-208 (^{208}Pb), the most abundant isotope of naturally occurring lead. Some calculations even indicated that element 110 with 184 neutrons (^{294}110) should be the longest-lived with a half-life in the range of hundreds of thousands to a billion years as shown in the contour plots (Fiset and Nix 1972, Randrup *et al.* 1974) in FIGURE 2.

SUPERHEAVY ELEMENTS - FIGURE 2

FIGURE 2. Contour plots of predicted half-lives of SHEs as a function of proton and neutron number according to a) Fiset and Nix, 1972 and b) Randrup *et al.*, 1974.

These elements near the predicted islands of nuclear stability around the spherical closed shells at proton numbers 110 to 114 (or even 126) and 184 neutrons are typically referred to as SHEs. Although arguments have been made (Armbruster and Münzenberg 1989) that the heavy elements that would not exist except for stabilization by nuclear shells, whether or not they are spherical, should be designated as SHEs, the term has usually been reserved for those elements in the region of the predicted spherical doubly magic nuclei.

A 1968 periodic table with the 'Superactinide' series proposed by Glenn T. Seaborg (1968) is shown in FIGURE 3. Seaborg stated that in placing what he called the 'Superactinides' at the bottom of the periodic table under the lanthanides and actinides, he had tried for simplicity's sake to conform as nearly as possible to the current form of the periodic table.

SUPERHEAVY ELEMENTS - FIGURE 3

FIGURE 3. Representation of Glenn T. Seaborg's 'conventional' form of the periodic table showing predicted location of new elements, including Superactinides, in parentheses.

This 32-member 'Superactinide' series would begin with element 122 and end with element 153 after filling of the $5g^{18}$ and $6f^{14}$ electronic shells, perhaps in mixed electronic configurations. However, Seaborg cautioned that complications from intruding 7d and 8p electrons might also occur and cause deviations from this picture and from the predominantly trivalent character expected for this Superactinide series. The predicted SHE with atomic number 126 would be a member of this series. A few examples of some of the early searches for SHEs in nature and at accelerators will be described in the next sections of this chapter and are discussed in much more detail by Hoffman, Ghiorso, and Seaborg in the Transuranium People: The Inside Story (Hoffman *et al.* 2000).

2. EARLY SEARCHES FOR SHES

2.1 In nature

Among the earliest searches for SHEs in nature were those conducted at Berkeley between 1968 and 1972 by S. G. Thompson's group (Nilsson *et al.* 1969b, Cheifitz *et al.* 1972). Searches were begun in natural ores for element 110 as eka-platinum and elements 111 through 114 as eka-gold, -mercury, -thallium, and -lead. Initially, they looked specifically for eka-platinum, element 110, using low background counting techniques and sensitive analytical methods. Among the most sensitive measurements are those based on evidence of SF decay. This process is very rare among the known naturally occurring radionuclides, and even if SHEs decay by other modes, they were expected to end in SF. The results of the searches were negative, corresponding to a concentration of $< 10^{-11}$ g/g. Later, they used their very high efficiency, large liquid scintillator system in an attempt to measure the high multiplicity of neutrons predicted (Nix 1969) to be emitted during the SF of spherical SHE nuclei in the region of element 114. In order to reduce effects from cosmic-ray background, they placed their detector some 250 m deep in a cross passage inside the Bay Area Rapid Transit (BART) system tunnel then under construction between Berkeley and Orinda, California. More than 40 large samples of ores, natural minerals such as galena and gold nuggets, manganese nodules from the ocean floor, moon rocks, and placer platinum were surveyed, but they found no evidence for increased neutron emission. Assuming a half-life of a billion years, they set a limit of $< 10^{-14}$ moles of SHEs per mole of sample.

In 1969, G. N. Flerov and co-workers (Flerov and Perelygin 1969) in Dubna, Russia reported detecting fission tracks in lead glass, which they attributed to the possible decay of SHEs. Based on additional observations of fission events in lead ore samples, they concluded they had found SF events with an apparent half-life of 4×10^{20} years that they attributed to SHEs. However, other researchers (Price *et al.* 1970) were not able to observe any fission tracks in old lead- and gold-rich minerals and set lower limits that were in contradiction with the Flerov results.

G. Herrmann (1974) published a review of early searches for SHEs in nature and concluded that no positive results had yet been obtained in either terrestrial or extraterrestrial samples, although many extremely sensitive methods had been developed and used in the search.

In 1976, R. V. Gentry and others (Gentry *et al.* 1976, Fox *et al.* 1976) reported evidence for element 126 and possibly elements 116, 124, and 127 in several 'Giant Halos' found in mica samples (see Section 13 of Chapter 6 in this Volume on pleochroic haloes). Gentry suggested that these giant halos had to be caused by 12 MeV to 14 MeV α particles, much higher than the known members of radioactive decay series, and might have been produced by SHEs. Measurements of miniscule samples containing a single giant halo were bombarded with low energy protons and the induced X-rays were measured. These so-called 'PIXIE' measurements indicated L X-rays at the appropriate energies for the SHEs 126 and possibly 116, 124, and 127 as well. These results created great excitement and elation in the community and, at first, it appeared that at last there was some firm physical evidence for SHEs. Unfortunately, the 'X-rays' turned out to be nuclear gamma rays induced in cerium-praseodymium isotopes present in the inclusions, and yet another discovery disappeared.

2.2 At accelerators

First attempts to produce SHEs 'artificially' were also conducted at Berkeley in 1968 by S. Thompson and A. Ghiorso and co-workers (Thompson 1968, Bowman *et al*. 1968). They used reactions of heavy ion projectiles with heavy actinide targets, e.g., $^{40}\mathrm{Ar} + {}^{248}\mathrm{Cm} \rightarrow {}^{288}114$, followed by neutron emission. No SHEs were detected and only limits could be set on the production cross sections and half-lives. (See remark #33 in Chapter 7 of Volume 1 on estimating such limits.)

A. Marinov *et al.* (1971a) published an article in *Nature* in January 1971, claiming production of element 112, eka-mercury, after they observed SF events in a mercury fraction chemically separated from the products of a long irradiation of tungsten with 24 GeV protons. The production mechanism was presumed to be secondary reactions with suitable targets of the heavy recoil products from the interactions of the high-energy protons. Later experiments showed (Marinov *et al.* 1971b) that some 70% of the observed

SF activity was due to contamination from ^{252}Cf, and subsequent attempts by members of the group to repeat the initial results were unsuccessful. Subsequent more sensitive experiments (Bimbot *et al.* 1971, Batty *et al.* 1973) also proved to be negative.

Flerov and Oganessian (1972) reported detecting SF activity with a half-life of about 150 days in sulfide fractions containing osmium and bismuth separated from the products of long bombardments of ^{238}U with a variety of ions as heavy as ^{136}Xe. However, the average number of neutrons per fission was typical of actinides rather than SHEs so, again, another report seemed unlikely. Attempts were initiated to try to produce SHEs in uranium + uranium collisions at the UNILAC in Darmstadt, Germany and in reactions of ^{48}Ca projectiles with ^{248}Cm at the SuperHILAC at Berkeley, but no positive results were obtained.

3. SUMMARY OF RESULTS TO 1978

The quest for SHEs continued in spite of the negative results, and in 1978 a large International Symposium on Superheavy Elements (Lohdi 1978) was held to assess the results and to consider future experiments. Most of the researchers working in the field and many other interested scientists attended. Some 15 countries and 50 different institutions were represented. The results of both searches in nature and attempts to synthesize SHEs were summarized. The quest had been extended by Anders and co-workers (Anders *et al.* 1975) to investigations of anomalous stable xenon ratios arising from fission of SHEs in meteorites, but the results were model dependent and controversial. The detection of neutrons from SF found in separated samples of hot springs water from the Cheleken Peninsula by Flerov and his group (Flerov 1977) was non-specific as pointed out by D. C. Hoffman (Hoffman 1978, Hoffman *et al.* 1980) and remained inconclusive. Investigations of the reaction of ^{48}Ca with ^{248}Cm at Berkeley (Hulet *et al.* 1977, Illige *et al.* 1978, Otto *et al.* 1978) continued to prove fruitless, and resulted only in pushing the limit on the production cross section still lower to 0.1 nanobarn (nb) (10^{-34} cm^2). In the closing summary of the Superheavy Elements Symposium, G. A. Cowan (1978), suggested using uranium beams to bombard ^{248}Cm, or even ^{250}Cm, recovered from underground nuclear tests. The conclusion was that there was still no positive evidence for the discovery of SHEs either in nature or in the products of accelerator bombardments. With the reduction of most of the half-life predictions from 10^9 down to 10^4 years, or even only a year depending on estimates of SF half-lives, most plans for future attempts to find SHEs were focused on production at accelerators.

4. SEARCHES FOR SHES SINCE 1978

In 1981 Herrmann summarized (Herrmann 1981) the results of searches for SHEs in damped collisions of ^{238}U with ^{238}U carried out at the UNILAC at Darmstadt utilizing their unique high-energy uranium beams to bombard uranium targets. A wide variety of radiochemical separation techniques and a rotating-wheel system were used to search for elements 108 to 118 and even 126. These experiments were also to no avail and upper limits of 10^{-32}, 10^{-33}, and 10^{-35} cm^2 were set on the cross sections for half-lives of 1 to 100 milliseconds, 100 milliseconds to 1 day, and 1 day to 1 year, respectively. However, he predicted that the planned increases in fluence levels would permit reaching cross sections below these levels. Subsequently, the search for the production of SHEs in damped collisions (see Chapter 3 in Volume 1) with ^{238}U (6.2-7.3 MeV/nucleon) at the UNILAC was extended to ^{248}Cm targets (Kratz *et al.* 1986) because this reaction was predicted to have a higher cross section. Comprehensive aqueous and gas-phase radiochemical separation techniques were used to isolate SHE fractions that were then assayed for SF activity, including fission-fragment total kinetic energies, and neutron emission. Again, the results were negative with upper cross-section limits of ~ 10^{-33} cm^2 for half-lives of minutes to hours and ~ 4×10^{-35} cm^2 for half-lives from days to several years.

About the same time, a large collaboration (Armbruster *et al.* 1985) of nuclear scientists, both chemists and physicists, from groups in the USA, Germany, and Switzerland conducted an exhaustive 'final' investigation of the reaction of ^{248}Cm with ^{48}Ca projectiles in 1982-83 first at the SuperHILAC at Berkeley, USA and then at the UNILAC in Darmstadt, Germany. The earlier attempts at Berkeley (Hulet *et al.* 1977, Otto *et al.* 1978, Illige *et al.* 1978) and Dubna (Oganessian *et al.* 1978) using bombarding energies corresponding to excitation energies of the compound system of 33 to 53 MeV had all been negative. Therefore, the new experiments were conducted with excitation energies between 16 and 40 MeV, close to the reaction barrier, in an attempt to keep the excitation energy as low as possible in order to minimize losses due to prompt fission.

The recoil fragment separators, Small Angle Separator System (SASSY) at the SuperHILAC, Lawrence Berkeley Laboratory (LBL) and Separator for Heavy Ion Reaction Products (SHIP) at the Universal Linear Accelerator (UNILAC) at the Gesellschaft für Schwerionenforschung (GSI), Darmstadt, Germany, were used to search for nuclides with half-lives as short as microseconds. In addition, both on-line and off-line radiochemical separation techniques were utilized to search for species with half-lives as short as a few seconds and as long as years. Again, no evidence was found for SHEs with production cross sections larger than 0.1 nb to 0.01 nb over a half-life range of 1 μs to 10 years.

Searches for naturally occurring SHEs in Atlantis II hot brine reported by Flerov and co-workers (Flerov *et al.* 1979), by Halperin and co-workers (Halperin *et al.* 1981), and by Feige and co-workers (Feige *et al.* 1987) also all proved to be negative. By the end of 1987,

no credible evidence for SHEs, either in nature or artificially produced, remained and the quest was essentially abandoned.

5. DISCOVERY OF ELEMENTS 107 THROUGH 112

Meanwhile, between 1981 and 1984, three new elements, bohrium (107), hassium (108), and meitnerium (109), were discovered (See Chapter 8 in this Volume.) A timeline of the discovery of the transuranium elements is shown in FIGURE 4. They were produced (Münzenberg *et al.* 1981, 1982a, b) at the UNILAC using so-called 'cold fusion' production reactions suggested by Oganessian *et al.* (1975). Targets of doubly magic stable ^{208}Pb or nearby stable ^{209}Bi were bombarded with the appropriate heavy-ion projectiles (e.g. enriched stable ^{54}Cr and ^{58}Fe ions). These 'shell-stabilized' targets react with the stable projectiles to give a compound nucleus that is the sum of their proton and neutron numbers. The resulting compound nuclei are produced with much lower excitation energies than those resulting from 'hot fusion' reactions in which unstable heavy actinide targets are used. These 'cold' compound nuclei are much more likely to de-excite by emitting only a single neutron, and thus, being less likely to be destroyed by fission, they have larger production cross sections. The isotopes ^{262}107, ^{265}108, and ^{266}109 were separated and identified using the in-flight Separator for Heavy-Ion reaction Products (SHIP), built at GSI under the direction of Peter Armbruster. The names and symbols bohrium (Bh) for 107, hassium (Hs) for 108, and meitnerium (Mt) for 109 were officially adopted for these elements along with rutherfordium (Rf) for element 104, dubnium (Db), formerly called hahnium (Ha) for element 105, and seaborgium (Sg) for 106 by the International Union of Pure and Applied Chemistry in August 1997 (CNIC 1997).

SUPERHEAVY ELEMENTS - FIGURE 4

FIGURE 4. Timeline of discovery of transuranium elements. The claimed discovery date is indicated rather than the date discovery was confirmed and approved by the IUPAC. Solid lines indicate confirmed elements, dashed lines indicate unconfirmed elements.

Evidence for different isotopes of element 110 was reported by several groups of scientists in 1995 to 1996. Ghiorso *et al.* (1995a, b) reported production of a single atom of ^{267}110 in the ^{209}Bi(^{59}Co,n) cold fusion reaction, Lazarev *et al.* (1996) reported evidence for decay of the neutron-rich nuclide ^{273}110 produced in the hot fusion reaction ^{244}Pu(^{34}S,5n), and Hofmann *et al.* (1995a) reported evidence for isotopes of element 110 produced in cold fusion reactions. More details on these reports are given in Hoffman (1998).

After many improvements to SHIP, an international team led by S. Hofmann conducted experiments at GSI in 1995 and claimed discovery of element 110 (Hofmann *et al*. 1995a) produced in the cold-fusion reaction ^{208}Pb(^{62}Ni, n) ^{269}110. They reported observation of ^{269}110 based on four chains that decayed by alpha emission to known daughter isotopes. They proposed the name darmstadtium (Ds) which was approved by the IUPAC in August 2003. The second chain could not be found in subsequent re-examination of the data (Hofmann *et al*. 2002), but IUPAC ruled that the remaining three chains constituted adequate proof.

In the subsequent experiments conducted in 1995, discovery of element 111 produced via the cold fusion reaction ^{209}Bi(^{64}Ni, n)^{272}111 was also claimed (Hofmann *et al*. 1995b) based on observation of 3 decay chains identified using SHIP. The name "roentgenium" (Rg) in honor of Wilhelm Conrad Roentgen was proposed for element 111 and announced on the 100th Anniversary of Roentgen's 1895 discovery of X-rays. The IUPAC considered the data of high quality but inconclusive because no known daughters were identified in the reported decay chains.

In 2004, K. Morita *et al.* (2004a) used the same reaction as the GSI group to produce ^{272}111. They observed 14 alpha-decay chains in total, thus providing much new information about its decay properties, clearing up some of the ambiguities and confirming the discovery of ^{272}111 and the α-decay properties previously reported by Hofmann *et al.* (1995b, 2002). The name "roentgenium" with symbol Rg was finally officially approved by IUPAC in November 2004.

Continuing their experiments at GSI using the SHIP separator, Hofmann *et al*. (1996) identified two decay chains attributed to element 112 produced via the ^{208}Pb(^{70}Zn,n)^{277}112 reaction. Hofmann *et al*. (2002) later reanalyzed their 1996 data and subsequently reported that one of the decay chains attributed to element 112 could not be found in the original data and performed additional experiments to obtain more data for the isotopes of element 110 through 112 to support their originally reported discoveries. Again, Morita *et al*. (2005, 2007a) at RIKEN repeated the GSI experiments on production of element 112. Their observation of two of the same decay chains as reported by the GSI team was important in the ultimate assignment of discovery of element 112 to the GSI team of Hofmann *et al*. (2002).

A Joint Working Party (JWP) appointed by the International Unions of Pure and Applied Chemistry (IUPAC) and Pure and Applied Physics (IUPAP) assigned priority of discovery in May 2009 of element 112 to the international group of scientists led by S. Hofmann (Hofmann *et al.*, 1996; Hofmann *et al.*, 2002) working at GSI. The discoverers proposed the name 'copernicium' with the symbol 'Cp' after the Polish astronomer Nicolaus Copernicus. However, the proposed symbol was amended to be 'Cn' because Cp for 'Cassiopeium' had earlier been proposed for element 71. That discovery could not be confirmed and element 71 was later named lutetium by the acknowledged discoverers. The approval process was initiated by IUPAC in July 2009 and probably cannot be completed

before mid-2010. The names and symbols for the transactinides approved as of 2009 are shown in TABLE 1. The updated 2009 Periodic Table of the Elements is shown in FIGURE 5.

TABLE 1. List of IUPAC approved transactinides (TANs) 104-112. (Elements 104-109 approved August 30, 1997).

Z	Name	Symbol
104	Rutherfordium	Rf
105	Dubnium (Hahnium)[#]	Db (Ha)[#]
106	Seaborgium	Sg
107	Bohrium	Bh
108	Hassium	Hs
109	Meitnerium	Mt
110[*]	Darmstadtium	Ds
111[**]	Roentgenium	Rg
112[***]	Copernicium[a]	Cn

[*]IUPAC approved August 2003.
[**]IUPAC approved November 2004.
[***]Discovery assigned May 2009.
[a]Required name and symbol comment period of more than 6 months initiated July 2009.

SUPERHEAVY ELEMENTS – FIGURE 5

FIGURE 5. Periodic table of the elements as of 2009.

One of the most significant things about the elements 107 through 112 is that they decay predominantly by α emission rather than SF, contrary to earlier predictions. These discoveries helped to give scientists renewed hope that it would be possible to reach the predicted island of SHE stability around element 114. The newly discovered nuclides also helped to substantiate the theoretical model and calculations of A. Sobiczewski and his group (Smolańczuk *et al.* 1995, Sobiczewski 1997) at the Soltan Institute for Nuclear Studies in Warsaw that predicted a doubly magic deformed region of extra stability around proton number 108 and neutron number 162 in addition to the island of spherical stability around $Z = 114$ and $N = 184$.

6. REPORTS OF DISCOVERIES OF SHES

6.1 1999-2003

After the discovery of element 112 in 1996, researchers at GSI attempted to produce element 113 but were unsuccessful. Extrapolation from their previous experiments with lead and bismuth targets led them to believe that the cross sections for producing the elements beyond 112 had dropped so low that they needed to further upgrade and increase the efficiency of their SHIP system before continuing the search.

Bombardment of ^{208}Pb with 449 MeV ^{86}Kr projectiles to produce ^{293}118 by a 1n out reaction was predicted by Smolańczuk (1999a, b) to have a large production cross section and decay via a unique chain of six high-energy α emitters with rather short half-lives. In 1999, the reaction was investigated by researchers at LBNL using the recently completed Berkeley Gas-filled Separator (BGS) at the 88-Inch Cyclotron. Because of the odd neutron in these nuclides, the half-lives might be as much as a factor of ten longer than these predictions. Due to the very low excitation energy of only 13.3 MeV calculated for the compound nucleus, emission of two neutrons is energetically forbidden, and single neutron emission is much more probable than alpha or proton emission from the compound nucleus. Ninov *et al.* (1999) reported finding three such decay chains with a cross section of a few picobarns in initial experiments conducted in April and May 1999, but upon re-examination of the original data, these results could not be verified. Later experiments conducted in 2001 (Gregorich *et al.* 2003) showed no evidence for these decay chains and an upper limit of about 1 pb for production of this decay chain in the reaction of ^{208}Pb with 449 MeV ^{86}Kr projectiles was set.

Concurrently, under the leadership of Yu. Ts. Oganessian, researchers from the Joint Institute of Nuclear Research (JINR), Dubna, Russia and the Lawrence Livermore National Laboratory (LLNL) heavy element group used the Dubna Gas-Filled Recoil Separator (DGFRS) to investigate production of heavier elements via the 'hot fusion' reaction. In bombardments of rotating ^{244}Pu targets with ^{48}Ca projectiles accelerated in the Dubna U-400 heavy ion cyclotron they found evidence for a single decay chain which they attributed to element 114 in data obtained from some 40 days of running time during November and December, 1998. This corresponds to a cross section of about 1 pb or less. Based on the bombarding energy of 236 MeV and the characteristics of the observed α-decay chain, they originally attributed the event to ^{289}114, which corresponded to the calculated maximum for the 3n evaporation reaction of the ^{244}Pu + ^{48}Ca reaction (Oganessian *et al.* 1999c). They reported observation of only one α-decay chain, a long chain ending with SF of ^{277}108. The measured time intervals between successive decays and the corresponding α-decay energies indicated relatively long half-lives for ^{289}114 (2-23 s), ^{285}112 (20-200 min), ^{281}110 (1-12 min), and ^{277}108 (16 min). The half-lives they reported for ^{285}112 and ^{281}110 are nearly a

million times longer than those of the heaviest previously known isotopes (^{277}112 and ^{273}110) of these elements. Because this reaction was not previously studied, the method of genetic correlations to known nuclei for the identification of the parent nucleus could not be used but the assignment of this one decay sequence to ^{289}114 seemed reasonable based on predicted excitation functions for the ^{244}Pu + ^{48}Ca reaction. Evidence for ^{288}114 via the 4n out reaction was subsequently observed in a continuation of the previous experiment that ran from June through October, 1999 using the same bombarding beam energy as the first experiment (Oganessian *et al.* 2000a.) Two decay sequences with similar decay characteristics were observed consisting of an implanted ion, two subsequent alpha-decays and terminated by a spontaneous fission. The decay chains were different than those observed during the first run. Based on previous experimental data as well as theoretical calculations, it was assumed that evaporation of either three of four neutrons from the ^{292}114 compound nucleus would be expected with similar probabilities (on order of one picobarn) over the range of excitation energies observed in both experiments, $E^* = 31.5 – 39$ MeV (Oganessian *et al.* 2000a). Therefore, observation of two neighboring isotopes of element 114 in these experiments at the same beam energy was not unexpected. The new chains obviously originated from a different parent nucleus than that previously observed during the first ^{244}Pu + ^{48}Ca reaction that were originally attributed to ^{289}114. The new nuclides observed in the second experiment were characterized by higher alpha-decay energies and terminated by SF at an earlier stage in the sequence. Comparison of the two chains supported assignment of the first reported decay chain (three alpha decays followed by SF) to the odd-mass nucleus ^{289}114 and the shorter chains consisting of two alpha decays followed by SF to even-even ^{288}114 (Oganessian *et al*., 2000a). However, in further measurements of the ^{244}Pu + ^{48}Ca excitation function over a range of higher beam energies (see Section 6.2), the one event originally attributed to ^{289}114 could not be reproduced and that result has never been confirmed. The original assignment of ^{288}114 has since been changed to ^{289}114 based on excitation function data described in Section 6.2.

In mid-April 1999, a multinational collaboration, also led by Oganessian, reported indirect evidence for two events of ^{287}114 from the ^{242}Pu(^{48}Ca,3n) reaction using the Dubna electrostatic recoil vacuum separator VASSILLISSA. These results were published (Oganessian *et al.* 1999b) in July 1999. In one event, a 10.29 MeV α particle was followed 1.3 s later by SF, while in the other event an escape peak with an α energy of only 2.31 MeV followed by SF was detected. The SF lifetimes for the two events were 9.3 min and 3.8 min, which the authors claim correspond to the same SF activity (within the limits of the reported uncertainties) of about 1.4 min that they produced previously in the reaction of ^{238}U with ^{48}Ca projectiles (Oganessian *et al.* 1999a) and had attributed to ^{283}112. In these experiments with the ^{242}Pu target, it was postulated that ^{283}112 would be observed as the alpha-decay daughter of ^{287}114. Based on the two SF events observed in the ^{242}Pu + ^{48}Ca reaction and the two events attributed to ^{283}112 produced via the ^{238}U(^{48}Ca,3n) reaction, the SF half-life of ^{283}112 was estimated to be approximately 3 min (Oganessian *et al*., 1999b).

However, later measurements of the ^{242}Pu + ^{48}Ca excitation function showed that the reported decay properties of ^{287}114 were not correct and the parent 114 nucleus actually decays through a chain consisting of two alpha particles followed by SF with the much shorter SF half-life of only a few seconds (Oganessian *et al.*, 2004b). The shorter ^{283}112 half-life was later reproduced by Hofmann et al. (2007), which supported the revised ^{287}114 decay properties (see Section 6.2).

In 2000, the Dubna/LLNL collaboration reported evidence for the production of ^{292}116 in the ^{248}Cm(^{48}Ca,4n) reaction at a center-of-target beam energy of 240 MeV (Oganessian *et al.* 2000b, 2001). One decay chain was observed consisting of an evaporation residue followed by two alpha particles and partial energy deposition of a third alpha followed by a spontaneous fission. The half-life estimated from this one event was ~ 33 ms; a cross-section limit was not given as the experiment was in progress during the reported observation of this nuclide, but subsequent excitation function measurements indicate it was likely on the order of 1 pb (Oganessian *et al.*, 2004b). The daughter nuclides appeared to decay with the same properties as those observed in the second reported ^{244}Pu + ^{48}Ca experiment where the ^{288}114 assignment was originally proposed (Oganessian *et al*., 2000a). Therefore, this first reported isotope of element 116 was assigned to ^{292}116, but was later changed to ^{293}116 when it was realized that the assignments of the first two reported isotopes of element 114 were incorrect (see Section 6.2 for more details). The earlier experiments of Armbruster et al. (1985) described in Section 4 reported no evidence for production of SHEs with cross sections larger than 0.1 nb (100 pb) to 0.01 nb (10 pb) over a half-life range of μs to 10 years. However, these experiments did not have sufficient sensitivity to detect SHE nuclides at the picobarn cross section level and are not in conflict with the above results.

6.2 2004-2009

The complete excitation function of the ^{244}Pu + ^{48}Ca fusion reaction was not measured until 2004 by Oganessian *et al*. (2004a). The ^{244}Pu targets were bombarded with ^{48}Ca ions at beam energies ranging from 243 to 257 MeV; these were higher than those studied previously during the first two reported observations of element 114 (Oganessian *et al*. 1999c, 2000a). At a ^{48}Ca energy of 243 MeV, decay chains consisting of two consecutive alpha-decays followed by SF were observed and had identical decay properties to the two chains previously reported at the lower bombarding energy of 236 MeV that were originally assigned to ^{288}114 (Oganessian *et al*., 2000a). In addition, at all three of these higher bombarding energies, very short decay chains consisting of a single alpha-decay followed by SF were observed, and at the highest beam energy of 257 MeV, one chain was detected that included two sequential alpha-decays ending with a SF with decay properties that

differed from any of the previous observed chains (Oganessian *et al.*, 2004a). It was assumed that the longer decay chains observed at the lowest and highest beam energies belonged to odd-mass isotopes of 114 while the short alpha-SF chains originated from an even-even nucleus. Based on predicted excitation functions for the ^{244}Pu + ^{48}Ca reaction, it now appeared that the single chain observed at the highest bombarding energy was likely due to the previously unreported isotope ^{287}114 produced via the 5n evaporation channel and the alpha-SF chain, therefore, originated from ^{288}114 produced via the 4n channel. These assignments meant that the decay chains originally attributed to ^{288}114 (Oganessian *et al*., 2000a) were actually from a different isotope, and based on the measurement of the excitation function, the assignment was changed instead to the 3n evaporation product, ^{289}114, which would be consistent with a longer, odd-mass decay chain (Oganessian *et al*., 2004a). The single decay chain originally reported as ^{289}114 (Oganessian *et al.*, 1999c) was never duplicated experimentally, and based on the results subsequently reported by Oganessian *et al*. (2004a), was probably not due to an isotope of element 114. Maximum cross sections for the production of 114 isotopes via the ^{244}Pu + ^{48}Ca reaction were reported to be: 2 pb (3*n*), 5 pb (*4*n), and 1 pb (5*n*).

Likewise, the complete excitation function of the ^{242}Pu + ^{48}Ca reaction was subsequently measured in 2004 over a range of beam energies from 235 to 250 MeV (Oganessian *et al.* 2004b). During a subsequent experiment, the ^{238}U + ^{48}Ca reaction was also studied over a range of beam energies from 230 to 240 MeV in order to identify the potential alpha-decay daughter of ^{287}114, ^{283}112 (Oganessian *et al*., 2004b). 15 decay chains consisting of two alpha-decays followed by SF were observed at three beam energies (235, 238, and 244 MeV) during bombardment of ^{242}Pu with ^{48}Ca. The decay properties matched those reported as ^{287}114 produced via the ^{244}Pu(^{48}Ca,5n) reaction (Oganessian *et al*., 2004a). This assignment was also supported by the direct production of the ^{283}112 daughter via the ^{238}U(^{48}Ca,3n) reaction (Oganessian *et al.* 2004b). ^{283}112, produced both directly and as the daughter of ^{287}114, was observed to decay in both cases via alpha-decay with a half-life of 4 s followed by a very short SF (Oganessian *et al.*, 2004b). These results disagreed with the original report of ^{283}112 (Oganessian *et al.*, 1999b), which indicated that it decayed via SF with a lifetime on the order of several minutes. The alpha-decay of ^{283}112 was later confirmed by Hofmann *et al*. (2007) but the 3-min SF activity was not observed. Hofmann *et al*. (2007) measured two decay chains from the ^{238}U(^{48}Ca,3n) reaction confirming data previously reported for this isotope by Oganessian *et al*. (2004b). Two other events were observed consistent with a 50% SF branch for ^{283}112. Decay chains for the observed element 114 isotopes based on the original measurements of their direct production via ^{242,244}Pu + ^{48}Ca reactions with their current proposed isotopic assignments are shown in Figure 6.

SUPERHEAVY ELEMENTS - FIGURE 6

FIGURE 6. Decay chains for ^{286}114 (Oganessian *et al*. 2004b), ^{287}114 (Oganessian *et al*. 2004a), ^{288}114 (Oganessian *et al*. 2004a), and ^{289}114 (initially reported as ^{288}114 by Oganessian *et al*. 2000a, later changed to ^{289}114 in Oganessian *et al*. 2004a) as measured from the direct production reactions shown on the figure. Half-lives calculated from average lifetimes; observed alpha energies in MeV are given. Yellow squares indicate alpha decay and green squares indicate SF.

Evidence for ^{286}114 was first reported by Oganessian *et al*. (2004a) as the daughter of ^{290}116 produced in the ^{245}Cm(^{48}Ca,3n) reaction shown in Figure 7. The assignment was later substantiated in measurements of cross sections for the ^{48}Ca + ^{242}Pu reaction (Oganessian *et al*., 2004b). The maximum cross section measured for production of ^{286}114 is approximately 4.5 pb for the ^{242}Pu(^{48}Ca,4n) reaction.

Additional isotopes of element 116, ^{290}116 and ^{291}116, were observed in the reaction of ^{48}Ca with ^{245}Cm (Oganessian *et al*. 2004a) with cross sections of about 1 pb at a bombarding beam energy of 243 MeV. Once the final assignments of ^{288}114 and ^{289}114 were determined by Oganessian *et al.* (2004a) (see discussion above) it was realized that the daughter of what was originally believed to be ^{292}116 had decay properties that, in fact, matched those of ^{289}114 produced in the ^{244}Pu + ^{48}Ca reaction (Oganessian *et al*., 2004a). Therefore, the assignment of the element 116 isotope originally observed in the ^{248}Cm + ^{48}Ca reaction was changed to ^{293}116 and ^{292}116 was later reported in the measurement of the ^{248}Cm(^{48}Ca,4n) reaction at 247 MeV with a cross section of about 1 pb (Oganessian *et al*., 2004b). Decay chains for the four observed isotopes of element 116 based on the original measurements of their direct production via ^{245,248}Cm + ^{48}Ca reactions (^{290,291,292,293}116) with their current proposed isotopic assignments are shown in Figure 7.

SUPERHEAVY ELEMENTS - FIGURE 7

FIGURE 7. Decay chains for ^{290}116 (Oganessian *et al*. 2004a), ^{291}116 (Oganessian *et al*. 2004a), ^{292}116 (Oganessian *et al*. 2004b), and ^{293}116 (initially reported as ^{292}116 by Oganessian *et al*., 2000b, 2001, later changed to ^{293}116 in Oganessian *et al*. 2004a) as measured from the direct production reactions shown on the figure. Half-lives calculated from average lifetimes; observed alpha energies in MeV are given. Yellow squares indicate alpha decay and green squares indicate SF.

The next reported discovery of a SHE was in 2004 when the Dubna/LLNL collaboration reported the observation of element 115 and its decay daughter 113 for the first time (Oganessian *et al.* 2004c, 2005). Targets of ^{243}Am were bombarded with 248-MeV ^{48}Ca projectiles resulting in a total beam dose of 4.3 x 10^{18} ^{48}Ca ions. Three decay chains

consistent with the decay of $^{288}115$ and its daughter $^{284}113$ formed via the $^{243}Am(^{48}Ca,3n)$ reaction were reported resulting in a cross section of about 3 pb. One additional decay chain corresponding to the decay of $^{287}115$ and its daughter $^{283}113$ produced via the $^{243}Am(^{48}Ca,4n)$ reaction was observed at an increased beam energy of 253 MeV giving a production cross section of about 1 pb. (See FIGURE 8).

SUPERHEAVY ELEMENTS – FIGURE 8

FIGURE 8. Reported decay chains for $^{287,288}115$ (Oganessian *et al*., 2004c, 2005). $^{271}107$ was not detected (ND). Half-lives calculated from average lifetimes; observed alpha energies in MeV are given. Yellow squares indicate alpha decay and green squares indicate SF.

The long-lived (16 h) ^{268}Db decay daughter of $^{288}115$ (Oganessian *et al.*, 2004c) was later isolated via chemical methods (Dmitriev *et al.* 2005) during a subsequent ^{48}Ca bombardment of ^{243}Am. During this procedure, the reaction products that came from the ^{243}Am were caught in a copper block placed directly behind the target. The block was removed after 24 h of irradiation and the surface was removed using a microlathe. The copper was then dissolved in nitric acid and run through a column of Dowex 50x8 cation-exchange chromatography resin. The Group IV and V elements were eluted together with hydrofluoric acid (Dmitriev *et al.*, 2005, Oganessian *et al.*, 2005, Schumann *et al.*, 2005). The entire effluent was evaporated on a thin polyethylene foil for subsequent alpha and SF counting. The entire procedure from end of bombardment to the beginning of counting was approximately 2-3 hours (Oganessian *et al*., 2005). Fifteen spontaneous fission events were observed with a measured half-life of 32 h (Oganessian *et al*. 2005), which was consistent within experimental uncertainties with the 16 h half-life observed for ^{268}Db produced as a decay product of the reported $^{288}115$ chain shown in FIGURE 8 (Oganessian *et al*. 2004c). However, the chemical method used would not differentiate the Group IV from Group V elements (Schumann *et al*., 2005). Identification of the atomic number cannot be made via spontaneous fission alone, especially in a case where contamination from other elements may have been present. The cross section for the production of $^{288}115$ via the $^{243}Am(^{48}Ca,3n)$ reaction was determined during the off-line chemical separation to be approximately 4 pb based on the assumption that the 15 observed SF events were from ^{268}Db and not another fissioning nuclide present in the final chemical fraction (Oganessian *et al.* 2005). This cross section was consistent with the 2.7 pb cross section measured via direct production (Oganessian *et al*. 2004c).

In a subsequent experiment performed by the LLNL/Dubna collaboration (Stoyer *et al.*, 2007, Wilk *et al.*, 2008), five SF events were detected in an experiment in which the reaction products collected in the copper catcher block were chemically separated to first isolate the Group V elements from other interfering products, and then the Group V fraction

was further separated into Nb- and Ta-like fractions in order to evaluate whether the previously observed fission events were from ^{268}Db, which would be expected to behave as a Group V element. After removing the surface of the copper and dissolving it as before, the solution was sorbed on an inert Kel-F resin coated with methyl isobutyl ketone (MIBK). Several chemical fractions were collected by performing gradient elutions with varying concentrations of HCl and HF (Wilk *et al*., 2008). The Group IV and V elements were shown to be in different fractions based on the location of tracer radionuclides (^{89}Zr, ^{92m}Nb, ^{177}Ta, ^{175}Hf, ^{167}Tm, and ^{169}Yb) added prior to the start of chemistry (Stoyer *et al.*, 2007). The group V elements were further separated in Nb- and Ta-like fractions through the use of graded HCl / HF elutions followed by a warm water wash, which removed all of the tantalum (Wilk *et al.* 2008). All fractions were evaporated onto thin polyethylene foils for subsequent alpha and SF counting. The entire procedure from the end of irradiation to the beginning of counting took approximately two hours. The five observed SF events all appeared in the Ta fraction, suggesting that the fissions originated from a Group V element. The lifetimes of the fission events were between 16 and 37 h. When one incorporates all of the fission events from direct production (Oganessian *et al*. 2004c) and the two subsequent off-line chemistry experiments (Oganessian *et al*. 2005, Stoyer *et al*. 2007, Wilk *et al*. 2008), the half-life of ^{268}Db is 28 h (Stoyer *et al*. 2007). The chemistry experiments provide indirect evidence that the daughter activity may come from the same long-lived dubnium nuclide that is part of the element 115 decay chain (Figure 8). However, direct production will ultimately be required to confirm assignment of ^{288}115.

Direct production of two different isotopes of element 113 has also been reported. Morita *et al.* (2004b, 2007b) reported production of two events of ^{278}113 via the ^{209}Bi(^{70}Zn,n) cold fusion reaction with the extremely small production cross section of only about 31 fb in two separate experiments at a beam energy of 353 MeV at the Riken Linear Accelerator Facility. These were attributed to ^{278}113 based on alpha-decay to the previously reported isotopes ^{266}Bh (approximately 1 s) (Wilk *et al*., 2000) and ^{262}Db (34 s). Oganessian *et al.* (2007a) reported observation of two decay chains of ^{282}113 produced via the ^{237}Np(^{48}Ca,3n) reaction at a bombarding energy of 244 MeV, corresponding to a production cross section of 0.9 pb. Decay chains for these experiments to produce isotopes of 113 are shown in Figure 9.

SUPERHEAVY ELEMENTS - FIGURE 9

FIGURE 9. Reported decay chains for Element 113. Left: ^{278}113, 2 events reported (Morita *et al.*, 2004b, 2007b). Right: ^{282}113 (two events, Oganessian *et al*., 2007a). Half-lives calculated from average lifetimes; observed alpha energies in MeV are given. Yellow squares indicate alpha decay and green squares indicate SF.

Eichler *et al.* (2007a, 2007b) studied the production of ^{287}114 via the ^{242}Pu(^{48}Ca,3n) reaction using thermochromatographic separation methods. The reaction products were volatilized and the ^{283}112 decay daughters were subsequently collected and measured on a detector surface cooled with a temperature gradient from +35°C to -186°C. The observed decay chains attributed to ^{283}112 (9.5 MeV alpha decay with a half-life of about 4 s) were consistent with those reported by Oganessian *et al*. (2004b) as decay products of ^{287}114 observed during measurement of the ^{242}Pu + ^{48}Ca excitation function (Oganessian *et al*,. 2004b). These experiments of Eichler *et al*. provide indirect proof of production of element 114 through the measurement of its ^{283}112 decay daughter.

Most recently, a group at Lawrence Berkeley National Laboratory (Stavsetra *et al.*, 2009) has reported production of one event each of ^{286}114 and ^{287}114 produced via the ^{242}Pu + ^{48}Ca reaction with decay properties consistent with those reported by Oganessian *et al*. (2004b). These experiments are the first to provide direct confirmation of production of element 114.

In 2006, Oganessian *et al*. (2006) reported the observation of three similar decay chains originating from element 118 produced via the ^{249}Cf(^{48}Ca,3n)^{294}118 reaction at two different excitation energies (29 and 34 MeV) with a cross section of approximately 0.5 pb. The assignment of element 118 to the new decay chains was supported by the genetic correlation with a previously identified isotope of element 116 measured in the ^{245}Cm(^{48}Ca,3n)^{290}116 reaction (Oganessian *et al*., 2004a) and its corresponding decay daughters. The decay chain for ^{294}118 is shown in Figure 10.

SUPERHEAVY ELEMENTS - FIGURE 10

FIGURE 10. Reported decay chain for ^{294}118 (Oganessian *et al*., 2006). Half-lives calculated from average lifetimes; observed alpha energies in MeV are given. Yellow squares indicate alpha decay and green squares indicate SF.

7. THEORETICAL PREDICTIONS FOR HALF-LIVES AND NUCLEAR PROPERTIES OF TRANSACTINIDE ELEMENTS (2003-2009).

Although most nuclear models indicated that half-lives and production rates rapidly decrease beyond Sg, some theoretical calculations (Sobiczewski 1997) predicted a region of nuclear stability around ^{270}Hs, a doubly magic deformed nucleus with 108 protons and 162 neutrons. One of the challenging experiments proposed for the future was to produce isotopes in this region for study of their properties. In recent studies by Dvorak *et al.* (2006, 2008), the new isotope ^{271}Hs (half-life ~4 sec) was produced via a 3n out reaction in the bombardment of ^{248}Cm with ^{26}Mg projectiles at below Coulomb barrier energies. The measured cross section of a few picobarns was comparable to the maximum cross sections for the previously known 4-s ^{269}Hs, as well as for the isotope ^{270}Hs which alpha-decays with a half-life of about 23 s and is an ideal candidate for chemical studies. These experiments suggest that the 3n reaction may be useful for making more neutron-rich transactinides using relatively light heavy ion beams and actinide targets.

These and other studies and reanalysis of data for Sg decay products of Hs alpha-decay by Düllmann and Türler (2008) have shown that the originally reported 21-s ^{266}Sg (Türler *et al.* 1998) was in error and was actually due to ^{265}Sg, which has two isomeric states with half-lives of 8.9 s and 16.2 s, respectively. The good news is that the 8.9-s ^{265}Sg isomer is produced preferentially via the ^{248}Cm(^{22}Ne,5n) reaction with an estimated cross section of a few hundred picobarns! Now it should be quite feasible to perform more "in-depth" detailed studies of the chemistry of Sg such as stability of its oxidation states both in solution and in gas-phase studies and complex formation under a variety of conditions.

A review of predicted chemical properties including ionization potentials, ionic radii, and redox potentials for Sg and lighter group 6 elements is given in Hoffman, Lee, and Pershina (2006). These indicate that the 6 oxidation state will be the most stable both in solution and in the gas phase and that Sg^{4+} will be even less stable than Ta^{3+} and W^{4+}. It will be especially interesting to compare actual comprehensive measurements of the chemical properties of Sg with those of lighter group 6 homologues and with the predictions of modern relativistic atomic and molecular calculations that include the influence of relativistic effects on chemical properties

The early predicted doubly magic spherical region of hypothetical long-lived SuperHeavy Elements (SHEs) shown in FIGURE 2 has never been reached although some of the recently reported heaviest isotopes of elements 112-118 produced in fusion reactions of heavy actinide nuclei with ^{48}Ca projectiles show significant increases in stability as the number of neutrons increases. Oganessian (2007b) has given a comprehensive review of the reported properties of the heaviest nuclides. He also discusses the theoretical predictions about the 'island of stability'.

Zagrebaev and Greiner (2008) considered the use of multinucleon transfer reactions in low-energy collisions of heavy ions for the production of new heavy isotopes along the closed shell at 126 neutrons which is important in the *r* process of nucleosynthesis. They predicted that more than 50 new nuclei might be produced in collisions of ^{136}Xe projectiles

with ^{208}Pb targets with cross sections of at least a microbarn and although SHEs would not be reached the information gained might be of some value in planning future SHE experiments and would add to our general information on reaction mechanisms.

Pei et al. (2009) investigated isentropic fission barriers using self-consistent nuclear density functional theory and the relationship between isothermal and isentropic descriptions. They found that the dependence of isentropic fission barriers on excitation energy changes rapidly and that shell effects are still important even for compound nuclei with large excitation energy. Fission barriers for nuclides produced in cold fusion reactions and those synthesized in hot fusion reactions are predicted to exhibit quite different behaviour. For nuclei around ^{278}112 produced in cold fusion reactions, they predict a more rapid decrease in the fission barriers with excitation energy compared to nuclei around ^{292}114 produced in hot fusion reactions. Research to provide reliable theoretical estimates of compound nucleus survivability is in progress.

In a 2005 review article, Ćwiok, Heenan, and Nazarewicz present new theoretical results for properties of even-even heavy and SHE element nuclei with $94 \leq Z \leq 128$ and with $134 \leq N \leq 188$. They use self-consistent formalism and a modern nuclear energy density functional to formulate the following major conclusions concerning SHEs: 1) SHE nuclei around Z=116 and N=176 are expected to exhibit coexistence of oblate and prolate shapes, in contrast to actinide and transfermium nuclei which have well-deformed elongated shapes; 2) Inclusion of triaxial shapes in the calculations dramatically reduces barriers between prolate and oblate shapes; 3) Gradual changes are expected along the triaxial energy surfaces although in special cases prolate and oblate shapes may be well separated. The heaviest isotopes recently reported by the Dubna/LLNL group are predicted to belong to a transition region that is subject to dramatic shape changes and/or triaxial softness. These shape effects are predicted to affect the alpha-decay energies, and half-lives may be lengthened due to hindrances in decay rates between parents and daughters with different shapes. Their predictions that the long-lived SHEs can exist in a variety of shapes and in some cases even in metastable states, and the formation of shape isomers will make positive identification of new species even more difficult than formerly anticipated.

8. FUTURE

The 1970s predictions of exceedingly long half-lives for nuclei in the region around the spherical doubly magic superheavy nucleus ^{298}114 have been drastically reduced. For example, calculations of Smolańczuk (1997) indicated that the spherical doubly magic superheavy nucleus ^{298}114 would decay predominantly by α–emission with a half-life of only about 12 min, but that ^{292}110 might α-decay with a half-life of about 50 years.

Theoretical calculations (Smolańczuk 2001, Chasman and Ahmad 1997) also indicated that nuclei with half-lives of microseconds or longer would exist all along the way to the predicted islands of stability. A 1978 contour plot with some of the nuclei reported in this 'transition' region as of mid-2002 is shown in FIGURE 11.

SUPERHEAVY ELEMENTS - FIGURE 11

FIGURE 11. Plot of heavy element topology from 1978 showing some landing points for proposed reactions. New heavy element isotopes reported as of mid-2002 are indicated with symbols denoting the following half-life ranges: + = 0.1 ms to 0.1 s; ο = 0.1 s to 5 min; • = > 5 min.

A large number of isotopes have been added to those shown as of mid-2002 for Sg (106) and beyond in Fig. 9 of our previous SHE paper (Hoffman and Lee 2003). Many assignments have changed, some have been dropped entirely, and a host of others have been added and still await confirmation. FIGURE 12 shows a similar representation of the isotopes *reported* (not necessarily confirmed) for Sg(106) through element 118 as of mid-2009 (Düllmann 2009).

SUPERHEAVY ELEMENTS - FIGURE 12

FIGURE 12. Chart of the Isotopes for Sg (106) through element 118 reported as of mid-2009 adapted from compilation of C. E. Düllmann, mid-2009 (Düllmann 2009).

Four isotopes each of elements 113 and 114, two of element 115, four of element 116, and one of element 118 have now been reported. It should be noted that none have yet been reported for element 117. Earlier calculations by Smolańczuk (1997) indicated that some of these nuclei would be expected to be nearly spherical with deformation energies ranging from only about 0.1 MeV for ^{292}116 to 0.2-0.3 MeV for the 114 isotopes, compared to zero deformation energy for the spherical doubly magic ^{298}114 and 7.8 MeV (Sobiczewski *et al.* 2001) for the doubly deformed magic nucleus, ^{270}108. Thus, the nuclei in this region may qualify as spherical SHEs even though they do not have the full complement of 184 neutrons.

A Dubna/LLNL team (Oganessian *et al.*, 2009) recently reported an unsuccessful attempt to synthesize element 120 at Dubna via the ^{244}Pu(^{58}Fe, xn)$^{302-x}$120 reaction. The sensitivity of the experiment corresponded to a cross section of ~0.4 pb for the detection of one decay. The production cross section of the evaporation residues in this reaction appeared to be lower by more than an order of magnitude compared to that of the same target with ^{48}Ca. They considered various explanations for this and concluded that

additional attempts to synthesize element 120 in this reaction would require much increased sensitivity and proposed that a more mass-asymmetric reaction, e.g., ^{248}Cm + ^{54}Cr or ^{249}Cf +^{50}Ti might be more favourable.

Subsequent to our mid-2002 review (Hoffman and Lee 2003), some 30 new heavy element isotopes have been reported. However, the longest lifetimes (not yet confirmed) reported to date by the Dubna/LLNL team for elements Rg through element 118 are: Rg-280: 3.6 s; 112–285, 29 s; 113–284, 0.5 s; 114–289, 2.6s; 115–288, 0.087 s; 116–293, 0.05 s; 117: not yet reported; 118–294, 0.09 ms. All of these decay via emission of an alpha particle. None are confirmed as yet and none decay to any currently known isotopes, although some of the results for production of element 114 have been duplicated by other groups. Making it still more difficult is that using currently known reactions, accelerators, and detection techniques only an atom every few days may be produced! The half-lives quoted above for Rg, 112, 114, and possibly 113 are long enough for chemical studies provided techniques can be developed to deal with or enhance the low production rates. On the positive side, it has already been demonstrated that it is possible to perform studies of chemical properties at the picobarn level for nuclides with half-lives of seconds. Perhaps some of the predicted longer-lived metastable states and shape isomers of SHEs can be produced with large enough production rates to permit identification and characterization of their nuclear decay properties. It has also been proposed (Armbruster 2008) that the magic proton shell should be shifted from Z=114 to Z=122, which would present an additional challenge for experimentalists to try to produce still more proton rich nuclei.

Although it now is predicted that many additional, relatively long-lived SHE species can exist, new production reactions, imaginative techniques for optimizing overall yields, and methods for 'stockpiling' long-lived products for off-line studies must be devised if these exciting new phenomena are to be fully explored.

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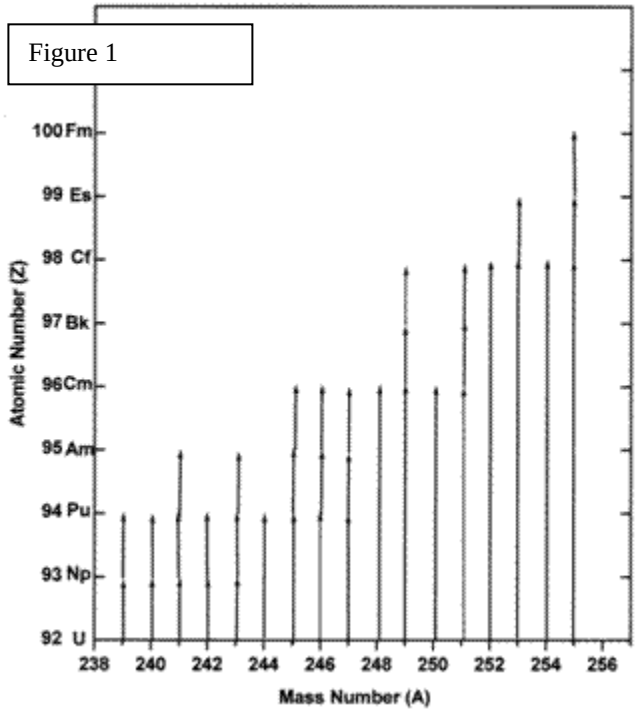
Figures are given below in order of figure number.

Figure 1



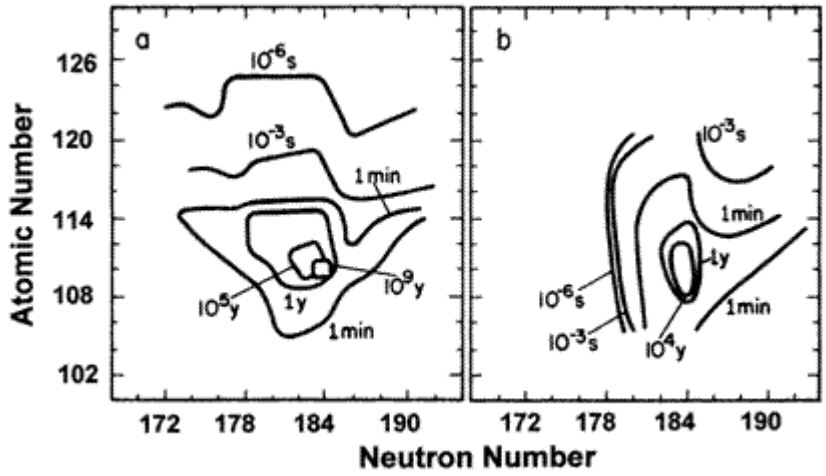


Figure 2

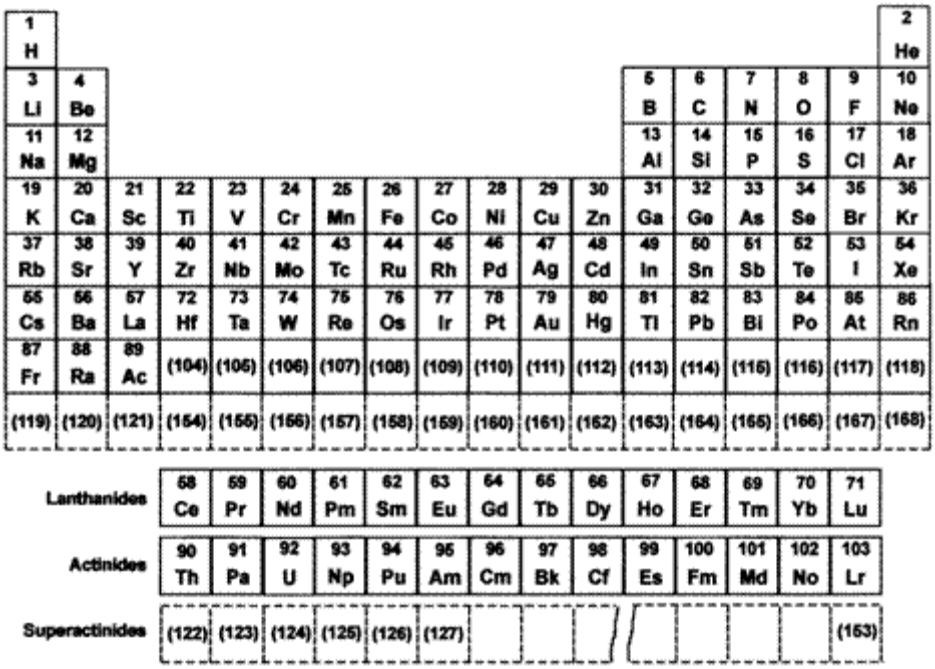


Figure 3

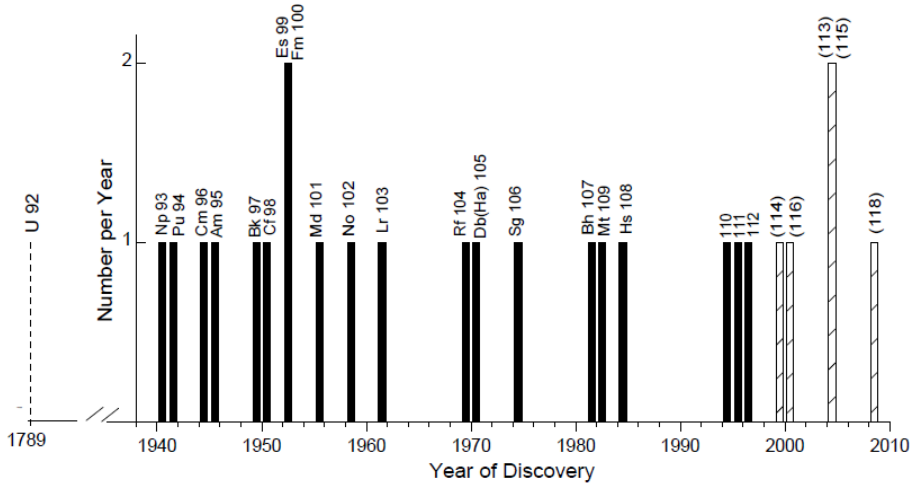


Figure 4

1																	18
1 H	2											13	14	15	16	17	2 He
3 Li	4 Be						Group					5 B	6 C	7 N	8 O	9 F	10 Ne
11 Na	12 Mg	3	4	5	6	7	8	9	10	11	12	13 Al	14 Si	15 P	16 S	17 Cl	18 Ar
19 K	20 Ca	21 Sc	22 Ti	23 V	24 Cr	25 Mn	26 Fe	27 Co	28 Ni	29 Cu	30 Zn	31 Ga	32 Ge	33 As	34 Se	35 Br	36 Kr
37 Rb	38 Sr	39 Y	40 Zr	41 Nb	42 Mo	43 Tc	44 Ru	45 Rh	46 Pd	47 Ag	48 Cd	49 In	50 Sn	51 Sb	52 Te	53 I	54 Xe
55 Cs	56 Ba	57 La	72 Hf	73 Ta	74 W	75 Re	76 Os	77 Ir	78 Pt	79 Au	80 Hg	81 Tl	82 Pb	83 Bi	84 Po	85 At	86 Rn
87 Fr	88 Ra	89 Ac	104 Rf	105 Db (Ha)	106 Sg	107 Bh	108 Hs	109 Mt	110 Ds	111 Rg	112 *(?)*	(113)	(114)	(115)	(116)		(118)

Lanthanides	58 Ce	59 Pr	60 Nd	61 Pm	62 Sm	63 Eu	64 Gd	65 Tb	66 Dy	67 Ho	68 Er	69 Tm	70 Yb	71 Lu
Actinides	90 Th	91 Pa	92 U	93 Np	94 Pu	95 Am	96 Cm	97 Bk	98 Cf	99 Es	100 Fm	101 Md	102 No	103 Lr

Figure 5

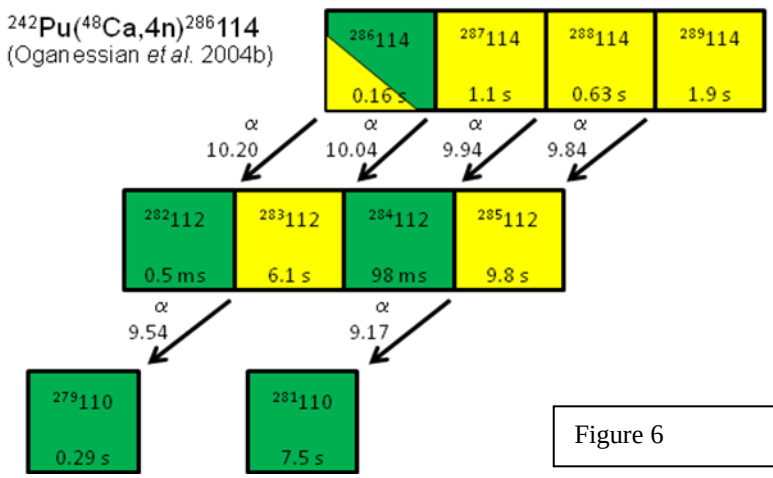


Figure 6

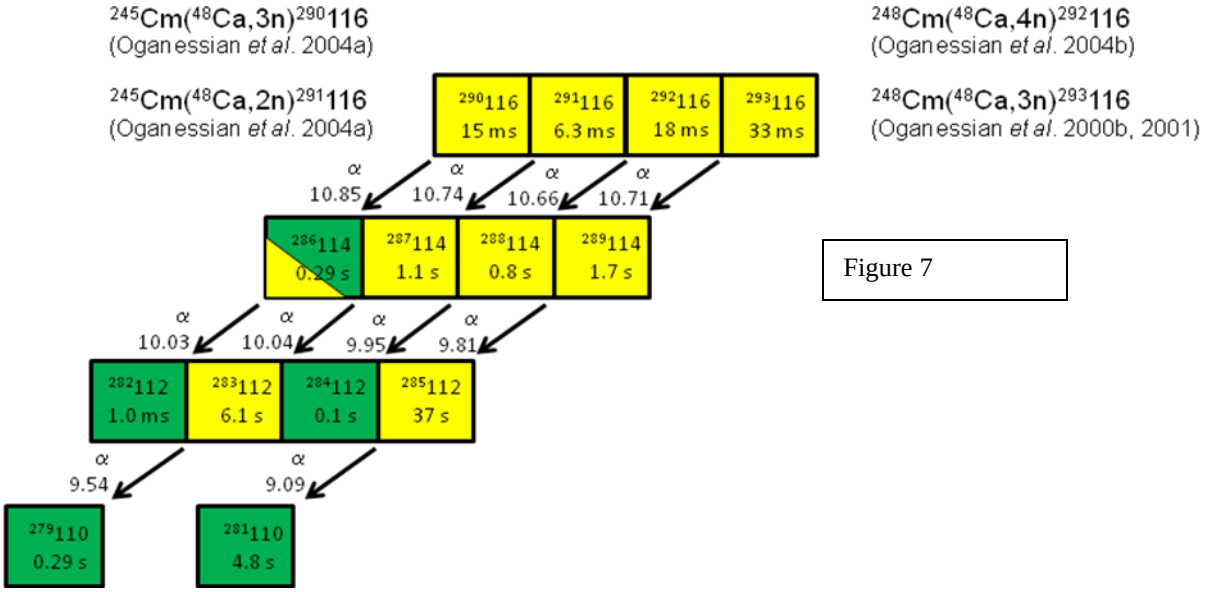


Figure 7

Figure 8

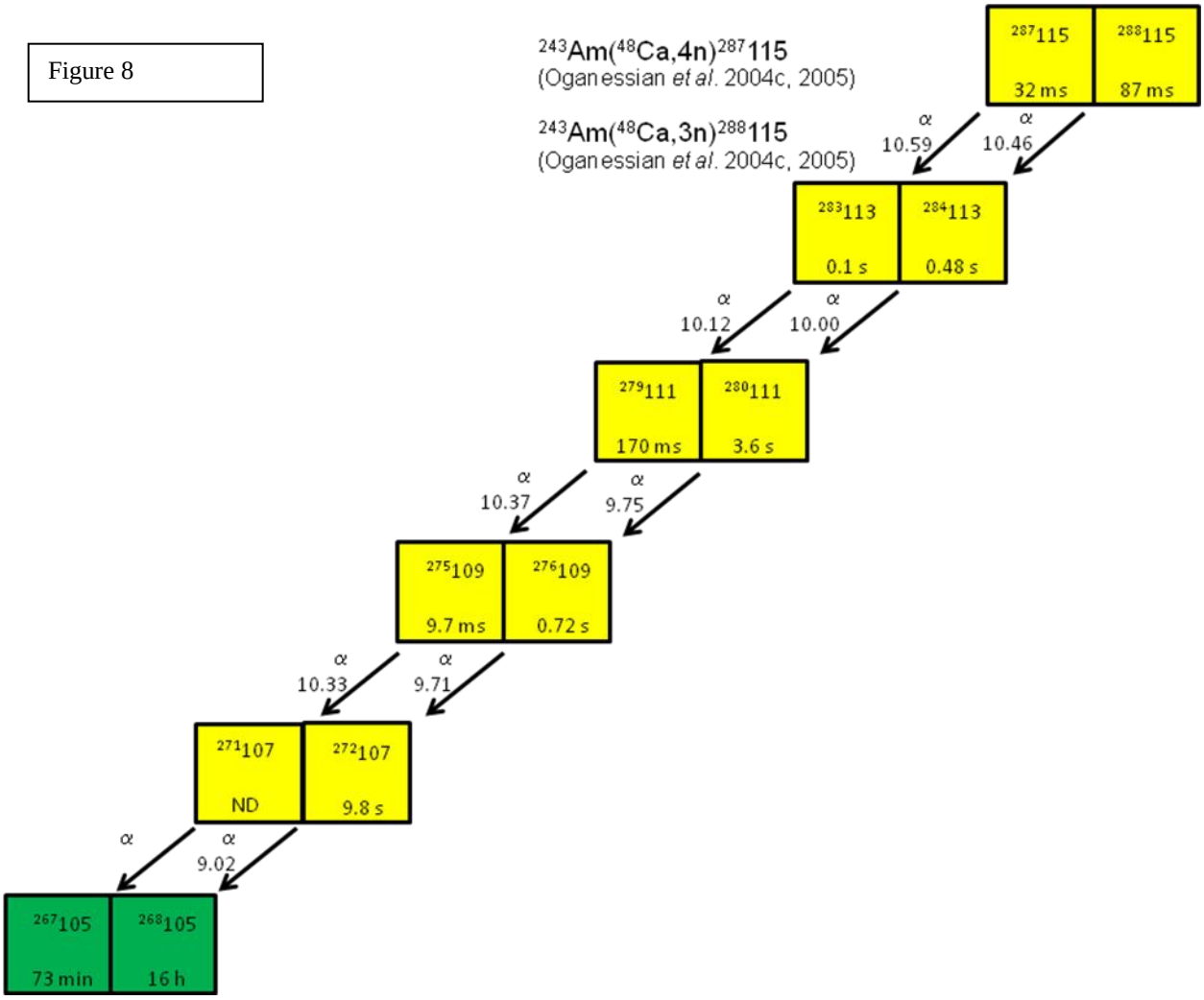
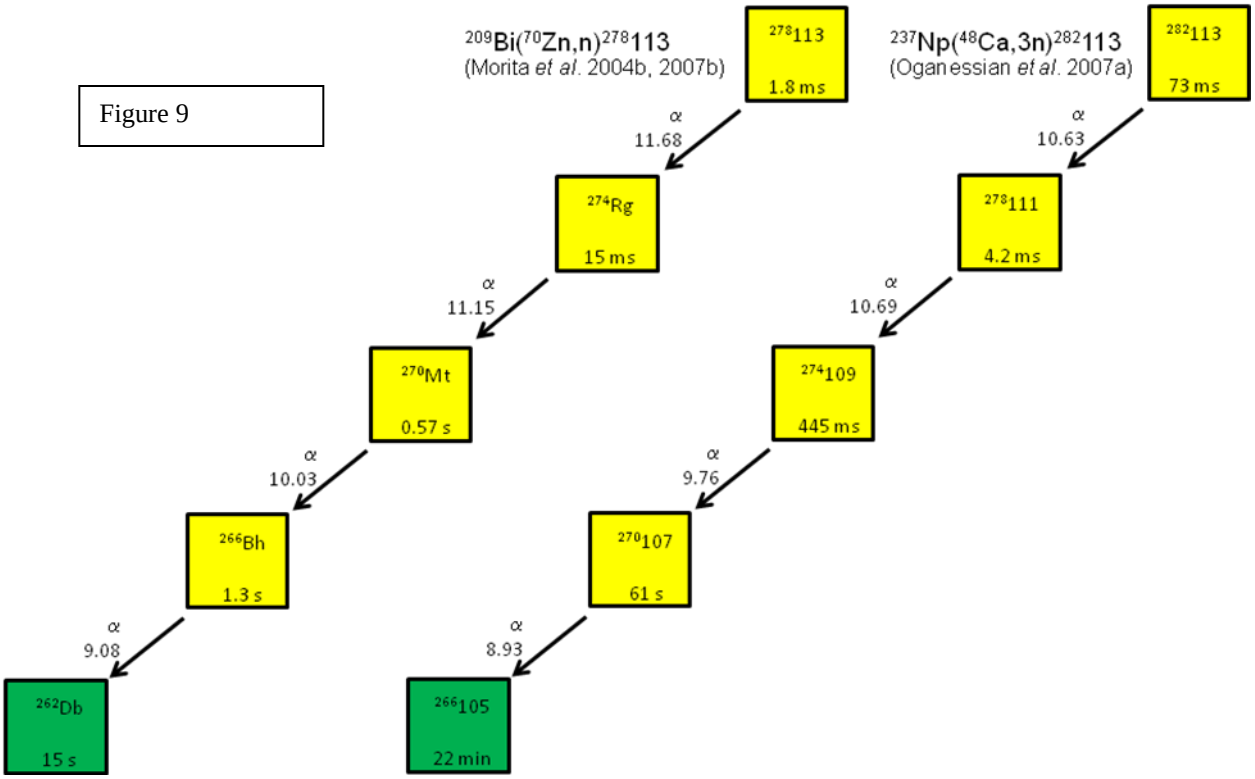

243Am(48Ca,4n)287115
(Oganessian et al. 2004c, 2005)
243Am(48Ca,3n)288115
(Oganessian et al. 2004c, 2005)
287115
32 ms
288115
87 ms
α 10.59
α 10.46
283113
0.1 s
284113
0.48 s
α 10.12
α 10.00
279111
170 ms
280111
3.6 s
α 10.37
α 9.75
275109
9.7 ms
276109
0.72 s
α 10.33
α 9.71
271107
ND
272107
9.8 s
α
α 9.02
267105
73 min
268105
16 h

Figure 9


209Bi(70Zn,n)278113
(Morita et al. 2004b, 2007b)
278113
1.8 ms
α
11.68
274Rg
15 ms
α
11.15
270Mt
0.57 s
α
10.03
266Bh
1.3 s
α
9.08
262Db
15 s
237Np(48Ca,3n)282113
(Oganessian et al. 2007a)
282113
73 ms
α
10.63
278111
4.2 ms
α
10.69
274109
445 ms
α
9.76
270107
61 s
α
8.93
266105
22 min

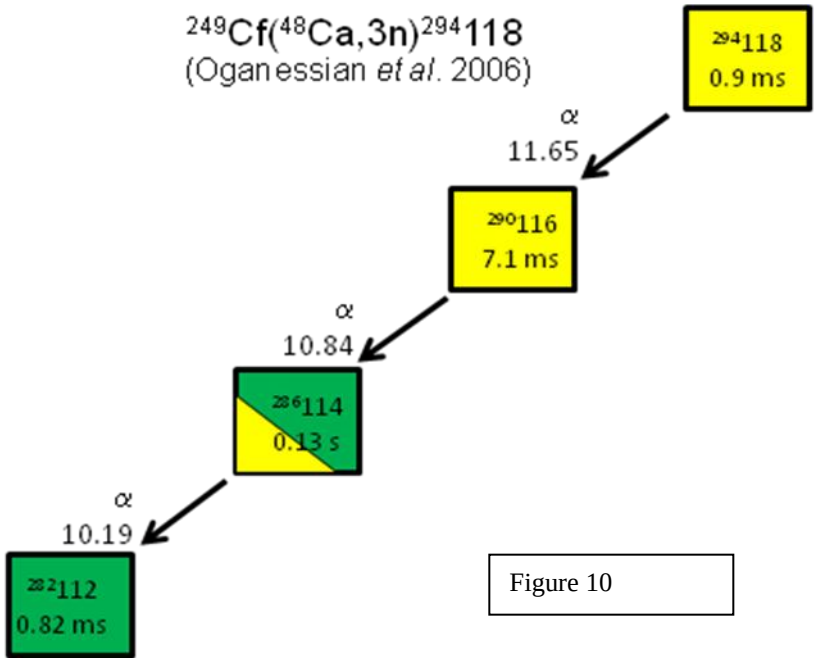

$^{249}Cf(^{48}Ca,3n)^{294}118$
(Oganessian *et al.* 2006)
$^{294}118$
0.9 ms
α
11.65
$^{290}116$
7.1 ms
α
10.84
$^{286}114$
0.13 s
α
10.19
$^{282}112$
0.82 ms

Figure 10

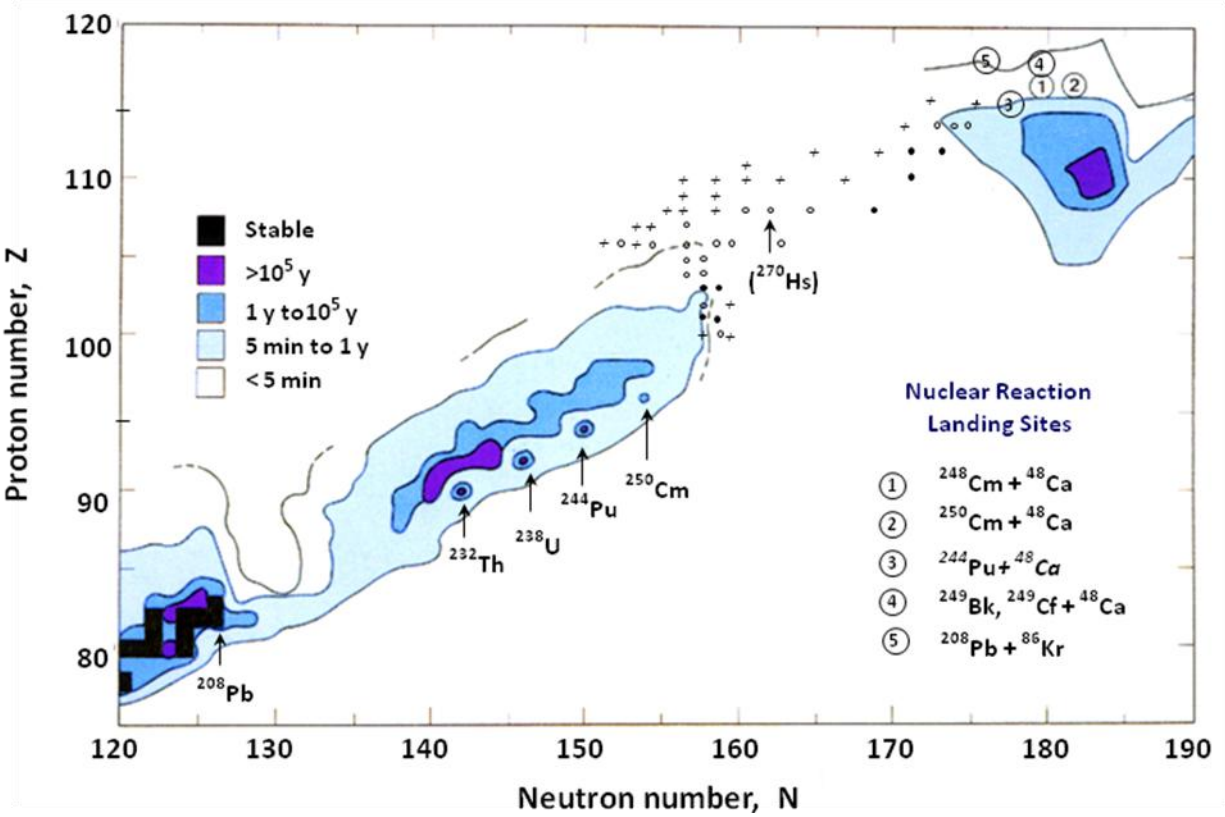
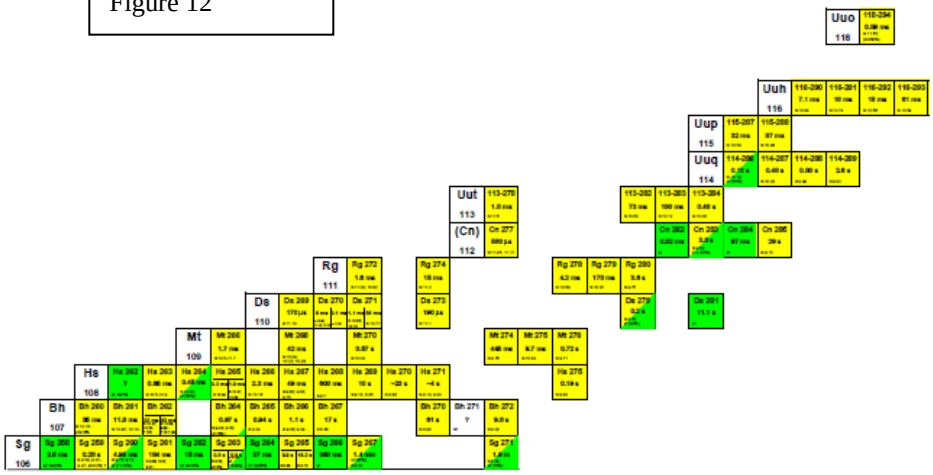

Proton number, Z
Neutron number, N
120
110
100
90
80
120
130
140
150
160
170
180
190
Stable
>10^5 y
1 y to10^5 y
5 min to 1 y
< 5 min
(270Hs)
250Cm
244Pu
238U
232Th
208Pb
Nuclear Reaction
Landing Sites
① 248Cm + 48Ca
② 250Cm + 48Ca
③ 244Pu+ 48Ca
④ 249Bk, 249Cf + 48Ca
⑤ 208Pb + 86Kr

Figure 11

Figure 12


Uuo
116
Uuh
116
Uup
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Uuq
114
Uut
113
(Cn)
112
Rg
111
Ds
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Mt
109
Hs
108
Bh
107
Sg
106